Challenges in modeling spatiotemporal phenomena in metal-organic frameworks

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Abstract

In this contribution, the question is addressed in how far current modeling strategies are capable of modeling spatiotemporal response in MOFs. The terminology spatiotemporal processes refers to the entanglement between the dynamics of the material and its spatial heterogeneities, where spatial heterogeneities from the subnanometer to the micrometer scale in real crystal particles with a finite size and morphology, impact the material's dynamics.1 Currently there exists a huge length-time scale gap between attainable theoretical length-time scales and experimentally relevant scales. Various challenges are ahead for modelers to bridge this length-time scale gap. First of all, systematic algorithms are needed that enable building atomistic models of realistic MOFs at the mesoscopic scale. Second methods are needed that allow evaluation of interatomic forces with quantum mechanical accuracy albeit at much lower computational cost than currently used Density Functional Theory methods. Third, methods are needed that allow to evaluate the kinetics of phenomena taking place in a multi length-time scale window to obtain an overall view of the dynamics of the process. With this contribution, I will highlight some of our recent efforts where we extended accessible length-time scales by exploited the massive parallelism of state-of-the-art GPUs using the OpenMM software package allowing to simulate MOFs having about a million number of atoms.2 The simulations necessitated to port our classical force fields to a GPU based framework and a new barostat had to be implemented that allowed to simulate anisotropic cell fluctuations. Despite the success of these simulations, showing how one can push the limits of currently accessible length and time scales, one loosed "quantum accuracy" when using classical force fields. Inspired by this defeat, we recently developed a new active learning based algorithm for developing machine learning potentials of activated processes.3 Within this framework a numerical potential is derived that can represent the interatomic forces with similar accuracy than the underlying quantum mechanical training data from which it was derived. This MLP based framework allows to simulate flexible behavior in MOFs in a much more efficient way. Our proof-of-concept results show that it is possible to train so-called universal MLPs which are viable for a class of materials. Furthermore, we have recently explored algorithms that allow to generate MLPs for defective materials by adding training data of some well chosen defective clusters. The illustrated methods show great potential to model spatiotemporal processes in realistic MOFs. As will become clear, to further progress in this field a close interaction loop with the experimental community on imaging, spectroscopy and characterization will be of utmost importance.